LA-UR- 10-00625

Approved for public release; distribution is unlimited.

Title: Measurement of Proton-Induced Delayed Neutron Cross-Sections ~~at 800 and 4000 MeV~~

Author(s): Randy J. Spaulding, Chris L. Morris, Steven J. Greene, Jeffrey D. Bacon, Gregory H. Canavan, Kiwhan Chung, Jay S. Elson, Gary E. Hogan, Mark F. Makela, Fesseha G. Mariam, Matthew M. Murray, Alexander Saunders, Zhehui Wang, Laurie S. Waters, Fred J. Wysocki

Intended for: APS "April" Meeting 2010
Feb 12-16, 2010
Washington, DC, USA

Los Alamos
NATIONAL LABORATORY
EST. 1943

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Form 836 (7/06)

Abstract Submitted
for the APR10 Meeting of
The American Physical Society

Sorting Category: E13. (E)

Measurement of Proton-Induced Delayed Neutron Cross-Sections ~~at 800 and 4000 MeV~~[1] RANDY SPAULDING, CHRIS MORRIS, STEVEN GREENE, GREG CANAVAN, ZHEHUI WANG, MARK MAKELA, LAURIE WATERS, KIWHAN CHUNG, FESSEHA MARIAM, JEFFERY BACON, Los Alamos National Laboratory, MATTHEW MURRAY, FRED WYSOCKI, GARY HOGAN, Los Alamos National Laboratory — Energetic proton beams are being used to explore delayed neutron emissions from a variety of materials in support of national security goals. To accomplish these goals it is necessary to have a firm understanding of the time structure of delayed neutrons emitted from the daughter nuclei of spallation, fragmentation, and fission interactions in structural and shielding materials as well as fissionable isotopes. Results of experiments conducted at Los Alamos National Laboratory (LANL) at 800-MeV and Brookhaven National Laboratory (BNL) at 4-GeV are presented.

[1]This work was supported by the Defense Threat Reduction Agency of the United States Department of Defense.

[X] Prefer Oral Session
[ ] Prefer Poster Session

Randy Spaulding
spaulding@lanl.gov
Los Alamos National Laboratory

Date submitted: 22 Oct 2009

Electronic form version 1.4

LA-UR- 09-07282

Measurement of Proton-Induced Delayed Neutron Cross-Sections a~~t 800 and 4000 MeV~~

List of Additional Authors:

Last	First	Middle	Z	Group
Morris	Christopher	L	081720	P-25
Greene	Steven	J	084378	P-25
Bacon	Jeffrey	D	190510	P-25
Canavan	Gregory	H	090633	P-DO
Chung	Kiwhan		188131	P-25
Elson	Jay	S	095755	D-5
Hogan	Gary	E	089987	P-25
Makela	Mark	F	153990	P-25
Mariam	Fesseha	G	094970	P-25
Murray	Matthew	M	083303	P-25
Saunders	Alexander		133587	P-25
Wang	Zhehui		151328	P-25
Waters	Laurie	S	108957	D-5
Wysocki	Fred	J	103123	P-24

MEASUREMENT OF PROTON-INDUCED DELAYED NEUTRON CROSS-SECTIONS

RANDY J. SPAULDING

P-25 THREAT REDUCTION TEAM
LOS ALAMOS NATIONAL LABORATORY

LA-UR-10-XXXXX

PRESENTED AT APS "APRIL 2010" MEETING
WASHINGTON, DC, 2/16/2010

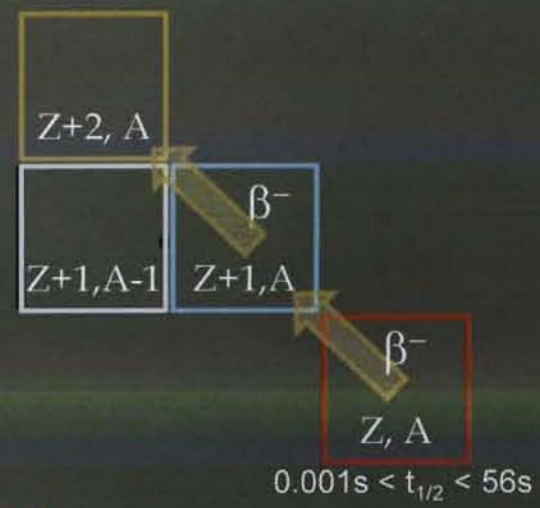
FUNDING FOR THIS WORK PROVIDED BY DTRA

Overview

- Beta-delayed neutron emitters
- Isotopic groups of emitters
- Surrogate method for scattered background subtraction
- Cross-Section data
  - 800-MeV (LANSCE)
  - 4-GeV (AGS)

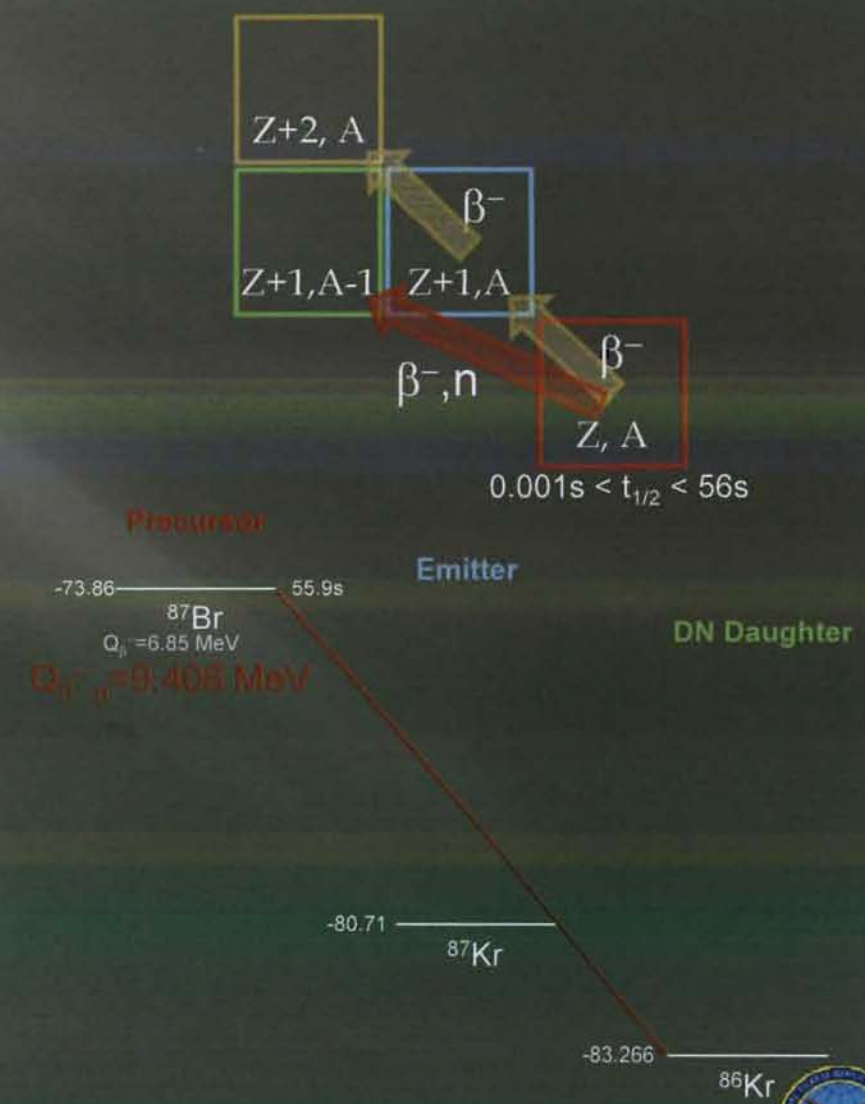
Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a large difference in excess mass between the two isobars.
  - $Q_{\beta^-} > 5$ MeV (typ.)



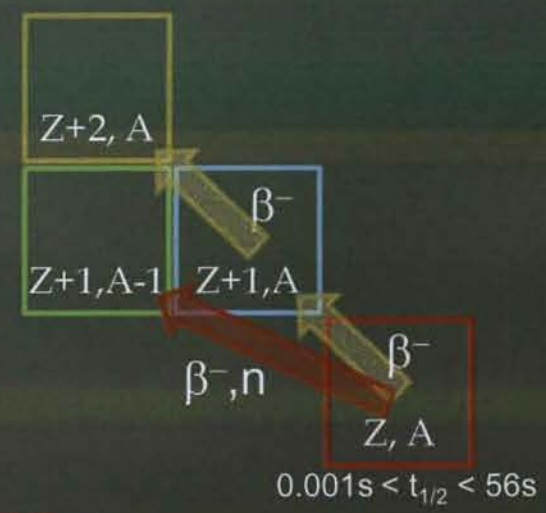
Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a large difference in excess mass between the two isobars.
  - $Q_{\beta^-} > 5$ MeV (typ.)
  - $Q_{\beta^-,n} > \Delta_n$ (8.071 MeV)

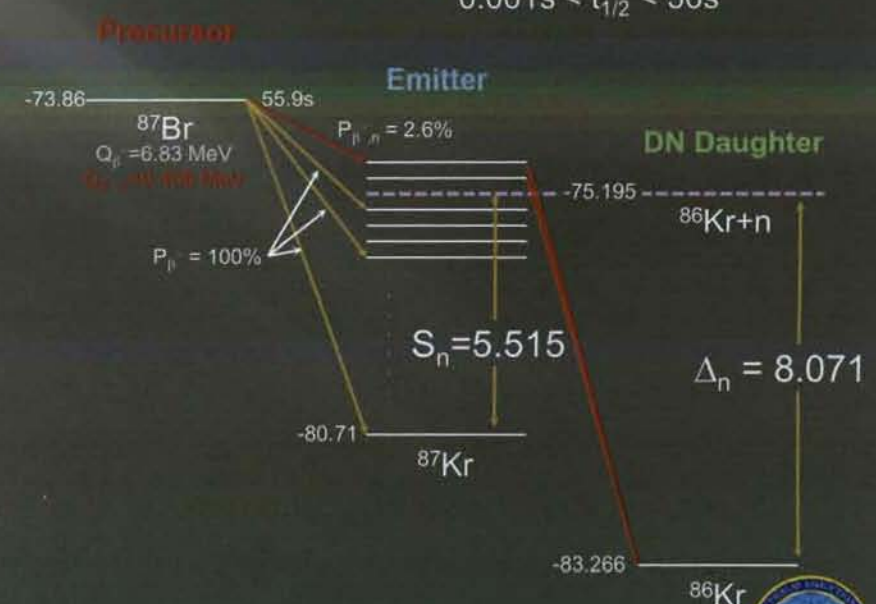


Beta-Delayed Neutron Production

- Isotopes that undergo delayed neutron emission following beta decay are characterized by a *large difference in excess mass* between the two isobars.
  - $Q_{\beta^-} > 5$ MeV (typ.)
  - $Q_{\beta^-,n} > \Delta_n$ (8.071 MeV)



- Lower-Z isobar (precursor) undergoes beta decay to an extremely short-lived excited state above the neutron binding energy of the emitter.



Delayed Neutron Precursors Appear in Clusters in Chart of Nuclides

Rb89	Rb90	Rb91	Rb92	Rb93	Rb94	Rb95	Rb96	Rb97	Rb98
Kr88	Kr89	Kr90	Kr91	Kr92	Kr93	Kr94	Kr95	Kr96	Kr97
Br87	Br88	Br89	Br90	Br91	Br92	Br93	Br94	Br95	Br96
Se86	Se87	Se88	Se89	Se90	Se91	Se92	Se93	Se94	
As85	As86	As87	As88	As89	As90	As91	As92		
Ge84	Ge85	Ge86	Ge87	Ge88					

Z

N

Clusters of Precursors Create Cascading Decay Chains

Rb89	Rb90	Rb91	Rb92	Rb93	Rb94	Rb95	Rb96	Rb97	Rb98
Kr88	Kr89	Kr90	Kr91	Kr92	Kr93	Kr94	Kr95	Kr96	Kr97
Br87 55.9 s	Br88	Br89	Br90	Br91	Br92	Br93	Br94	Br95	Br96
Se86	Se87 5.8 s	Se88 1.5 s	Se89	Se90	Se91	Se92	Se93	Se94	
As85	As86	As87 0.6 s	As88	As89	As90	As91	As92		
Ge84	Ge85	Ge86	Ge87	Ge88					

Z

N

Clusters of Precursors Create Cascading Decay Chains

Decay Chains are NOT Simple Exponential!

$$\begin{cases} \frac{d}{dt} Se87 = -\lambda_{Se87} \cdot Se87(t) \\ \frac{d}{dt} Se88 = -\lambda_{Se88} \cdot Se88(t) \\ \frac{d}{dt} Br87 = -\lambda_{Br87} \cdot Br87(t) + \lambda_{Se87} \cdot (1 - \beta_{Se87}) \cdot Se87(t) + \lambda_{Se88} \cdot \beta_{Se88} \cdot Se88(t) \end{cases}$$

where $\beta_i \equiv$ delayed neutron branching ratio

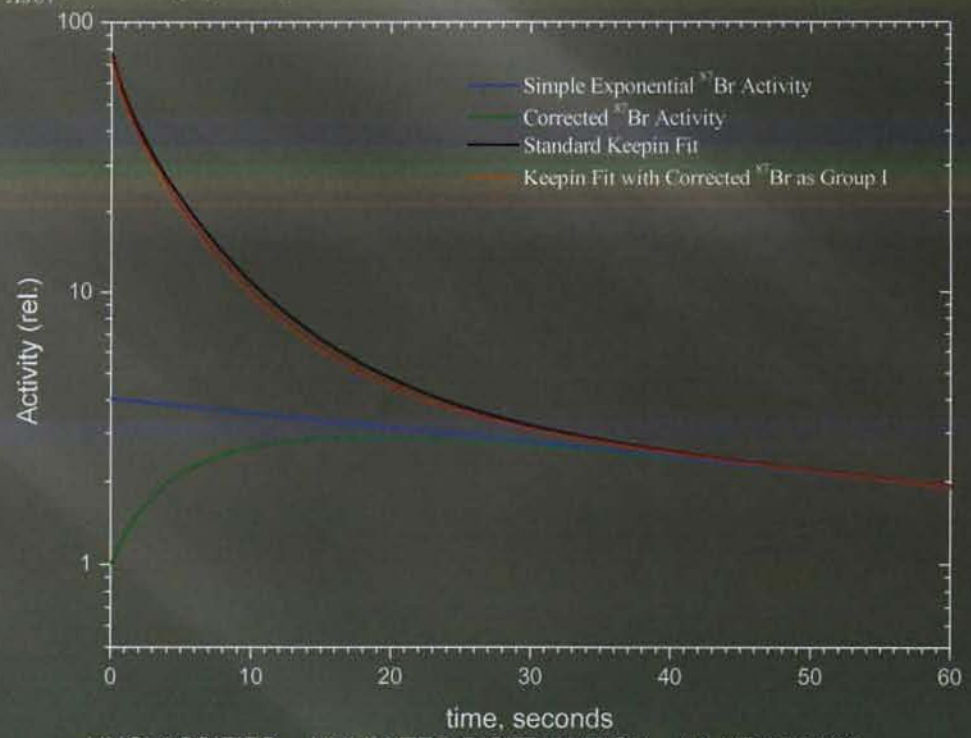
Decay of Beta Precursors is not Always Simple Exponential

$$\alpha_{Br87}(t) = \lambda_{Br87} \cdot \rho \cdot \left(\frac{Se87(0) \cdot \lambda_{Se87}}{\lambda_{Br87} - \lambda_{Se87}} \cdot (1 - \beta_{Se87}) \cdot e^{-\lambda_{Se87} \cdot t} + \frac{Se88(0) \cdot \lambda_{Se88}}{\lambda_{Br87} - \lambda_{Se88}} \cdot \beta_{Se88} \cdot e^{-\lambda_{Se88} \cdot t} + K \cdot e^{-\lambda_{Br87} \cdot t} \right)$$

where $\beta_i \equiv$ delayed neutron branching ratio

$\rho \equiv$ initial amount of each isotope created (appx. equal)

$Se87(0) = 3 - 2 \cdot \beta_{As87}$, $Se88(0) = 3$, $K = BUNC$



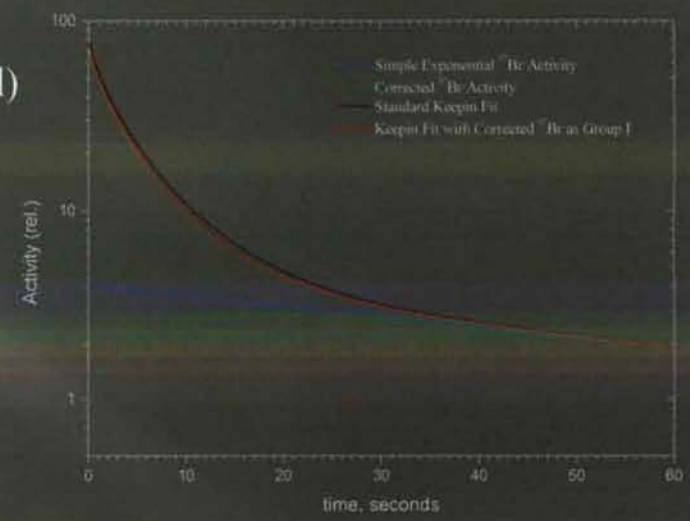
Decay of Beta Precursors is not Always Simple Exponential

$$\alpha_{Br87}(t) = \lambda_{Br87} \cdot \rho \cdot \left(\frac{Se87(0) \cdot \lambda_{Se87}}{\lambda_{Br87} - \lambda_{Se87}} \cdot (1 - \beta_{Se87}) \cdot e^{-\lambda_{Se87} \cdot t} + \frac{Se88(0) \cdot \lambda_{Se88}}{\lambda_{Br87} - \lambda_{Se88}} \cdot \beta_{Se88} \cdot e^{-\lambda_{Se88} \cdot t} + K \cdot e^{-\lambda_{Br87} \cdot t} \right)$$

where $\beta_i \equiv$ delayed neutron branching ratio

$\rho \equiv$ initial amount of each isotope created (appx. equal)

$Se87(0) = 3 - 2 \cdot \beta_{As87}$, $Se88(0) = 3$, $K = BUNC$



- Small effect on count rate (5-20%)
- Neglected in historical multigroup fits
- Should be negligible in this application

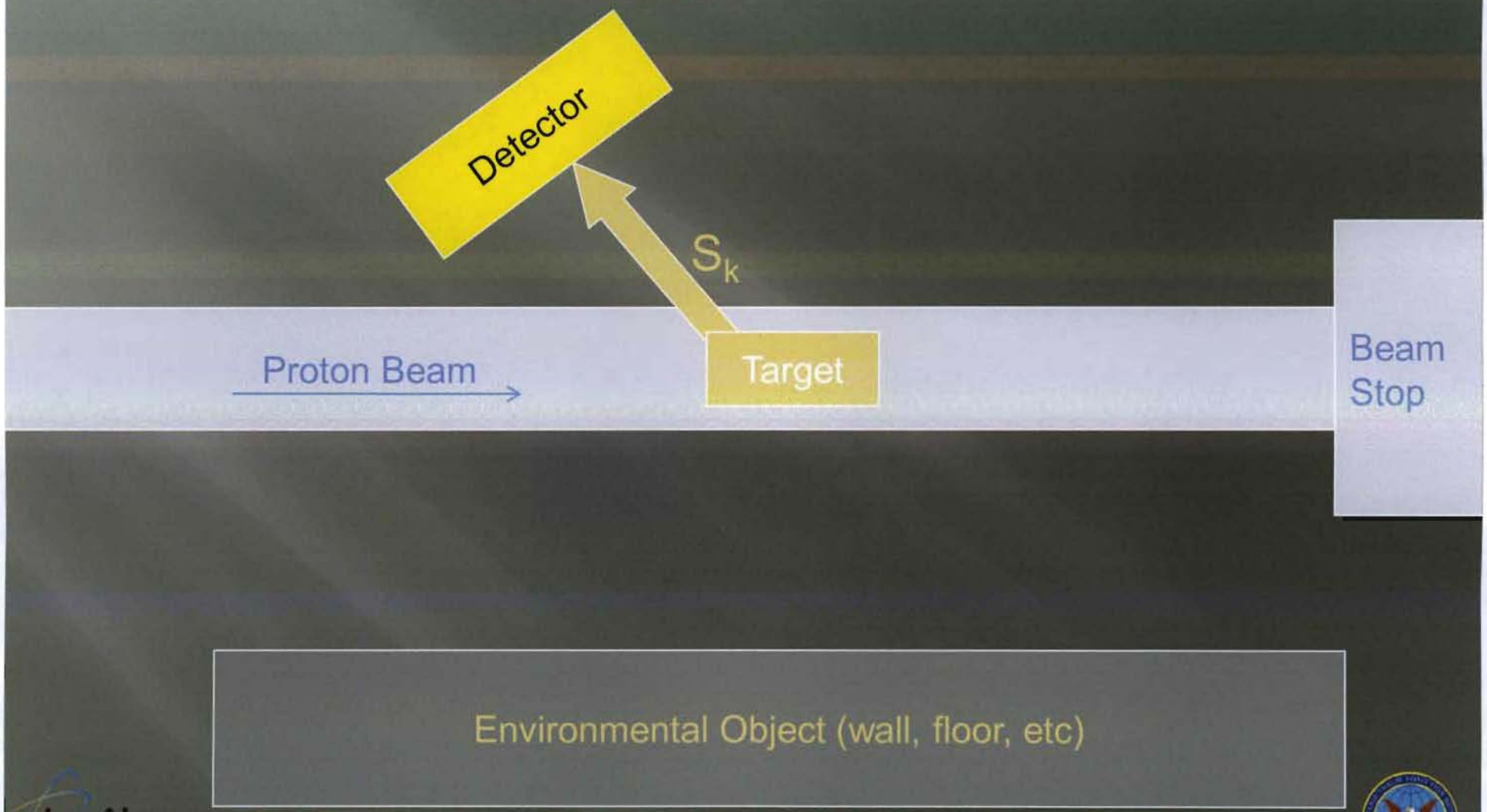
Decay Chains Can Be Quite Complex

Isotopic Target Group Definitions

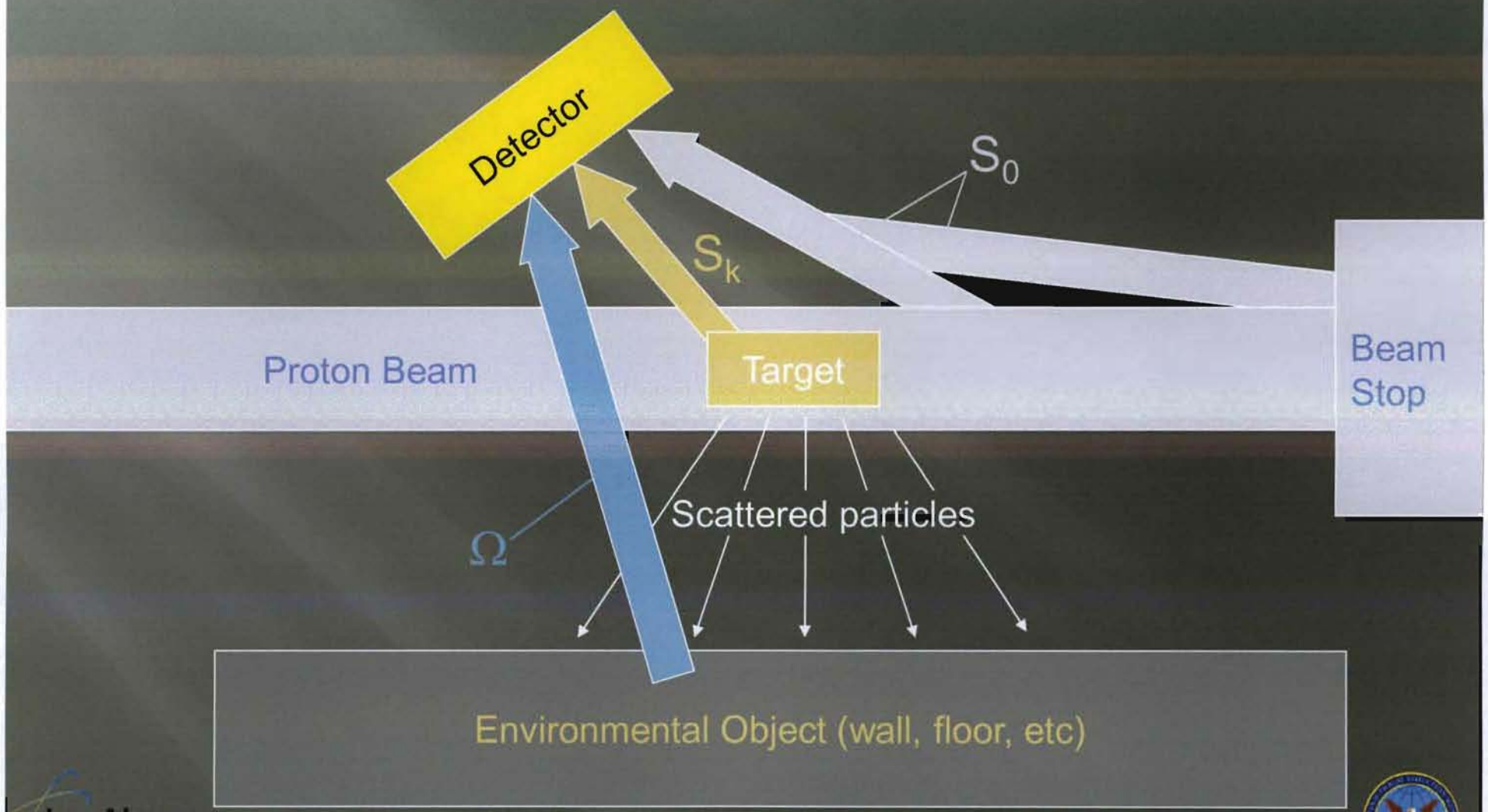
- Grouped by atomic number based on half-lives of beta precursors that can be produced

Group	Atomic Number	Decay Characteristics Example Precursors
0	A < 8	No beta precursors
1	9 <= A < 17	Large cross-sections, very short half-life ^{9}Li (178 ms)
18	^{18}O	Enormous (p, 2p) cross-section ^{17}N (4.17 s)
2	17 <= A < 50	Cross-section drops with A ^{17}N, ^{22}O (2.25s), ^{16}C (747 ms), ^{18}N (624 ms), ^{19}N (271 ms)
3	50 <= A < 90	Cross-section and time signature fairly constant ^{51}Ca (10s), ^{48}K (6.8s), ^{49}K (1.26s), ^{50}K (472 ms), ^{44}Cl (560 ms)
4	90 <= A < 209	Cross-section increases slowly with A, small long-time tail ^{87}Sr (55.65s), ^{137}I (24.5 s), ^{88}Br (16.29s), ^{85}As (2.02s), ^{91}Br (541 ms)
5	233 <= A	Large long-time tail, significant multiplication factor for fissiles

Determining Beam-Induced Background with a Surrogate Target



Determining Beam-Induced Background with a Surrogate Target



Count Rates in Surrogate Experiment

- C_k = Count rate for in-beam targets
- S_k = Signal from various targets:
- B = Non-beam background count rate
- Ω_k = Scattered background component
- p_k = Protons in beam pulse
- κ_k = Fraction of protons passing through target
- ε = Detector efficiency (time-dependent! But neglect for now)

$$S_k = \frac{\sigma_k \lambda_k N_A}{A_k}$$

$$C_k = \varepsilon p_k S_k$$

A_k = atomic number

λ_k = mass thickness in g/cm^2

Count Rates in Surrogate Experiment

- C_k = Count rate for in-beam targets
- S_k = Signal from various targets:
- B = Non-beam background count rate
- Ω_k = Scattered background component
- p_k = Protons in beam pulse
- κ_k = Fraction of protons passing through target
- ε = Detector efficiency (time-dependent! But neglect for now)

$$S_k = \frac{\sigma_k \lambda_k N_A}{A_k}$$

$$C_k = \varepsilon p_k S_k$$

A_k = atomic number

λ_k = mass thickness in g/cm^2

$$C_0 = \varepsilon \cdot \left(B + p_0 S_0 \right)$$

$$C_C = \varepsilon \cdot \left\{ B + p_C \left[S_0 + \kappa_C \left(S_C + \Omega_C \right) \right] \right\}$$

$$C_i = \varepsilon \cdot \left\{ B + p_i \left[S_0 + \kappa_i \left(S_i + \Omega_i \right) \right] \right\}$$

Subscripts: 0 = Null target (air column + beam stop)
C = Surrogate target (Carbon)
i = Various interrogation targets

Parameters and Count Rates in Surrogate Experiments

- C_k = Count rate for in-beam targets
- S_k = Signal from various targets:
- B = Non-beam background count rate
- Ω_k = Scattered background component
- p_k = Protons in beam pulse
- κ_k = Fraction of protons passing through target
- ε = Detector efficiency (time-dependent! But neglect for now)

$$S_k = \frac{\sigma_k \lambda_k N_A}{A_k}$$

$$C_k = \varepsilon p_k S_k$$

A_k = atomic number

λ_k = mass thickness in g/cm^2

$$C_0 = \varepsilon \cdot (B + p_0 S_0) \qquad \rightarrow S_0 = (C_0/\varepsilon - B)/p_0$$

$$C_C = \varepsilon \cdot \{B + p_C [S_0 + \kappa_C (S_C + \Omega_C)]\} \qquad \rightarrow \Omega_C = ((C_C/\varepsilon - B)/p_C - S_0)/\kappa_C$$

$$C_i = \varepsilon \cdot \{B + p_i [S_0 + \kappa_i (S_i + \Omega_i)]\}$$

Set surrogate signal to Zero!

Subscripts: 0 = Null target (air column + beam stop)
C = Surrogate target (Carbon)
i = Various interrogation targets

UNCLASSIFIED – UNLIMITED DISTRIBUTION – LA-UR-XXXXX

Surrogate Cross-Section Calculation

- Assume S_C=0 (no delayed neutrons from surrogate)
  - For Carbon surrogate this is valid for $t_D > 2s$ *
- Define $\eta_i \equiv \Omega_i / \Omega_C$ as the ratio of neutrons produced from beam scattered by the target divided by the neutrons produced from surrogate

$$S_i = \frac{1}{\varepsilon \kappa_i}\left\{ dC_i + \varepsilon B\left[\frac{1}{P_0} - \frac{1}{P_i} + \alpha_i\left(\frac{1}{P_C} - \frac{1}{P_0}\right)\right] + (\alpha_i - 1)\cdot dC_0 - \alpha_i \cdot dC_C \right\}$$

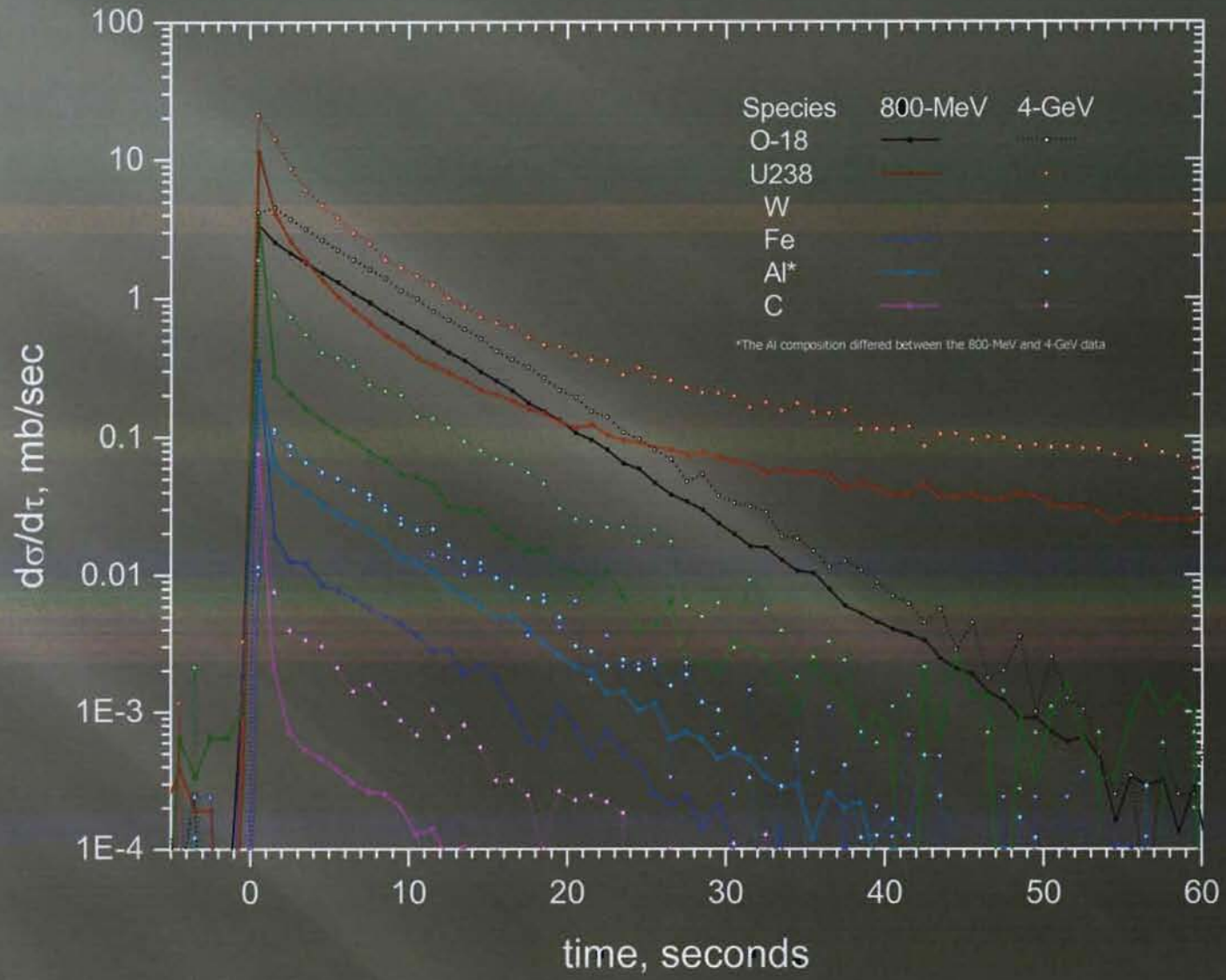
where $\alpha_i \equiv \eta_i \kappa_i / \kappa_C$, $dC_i \equiv C_i / p_i$.

* 2 seconds is roughly 10 half-lives of ^{9}Li, the longest-lived precursor produced from a Carbon target.

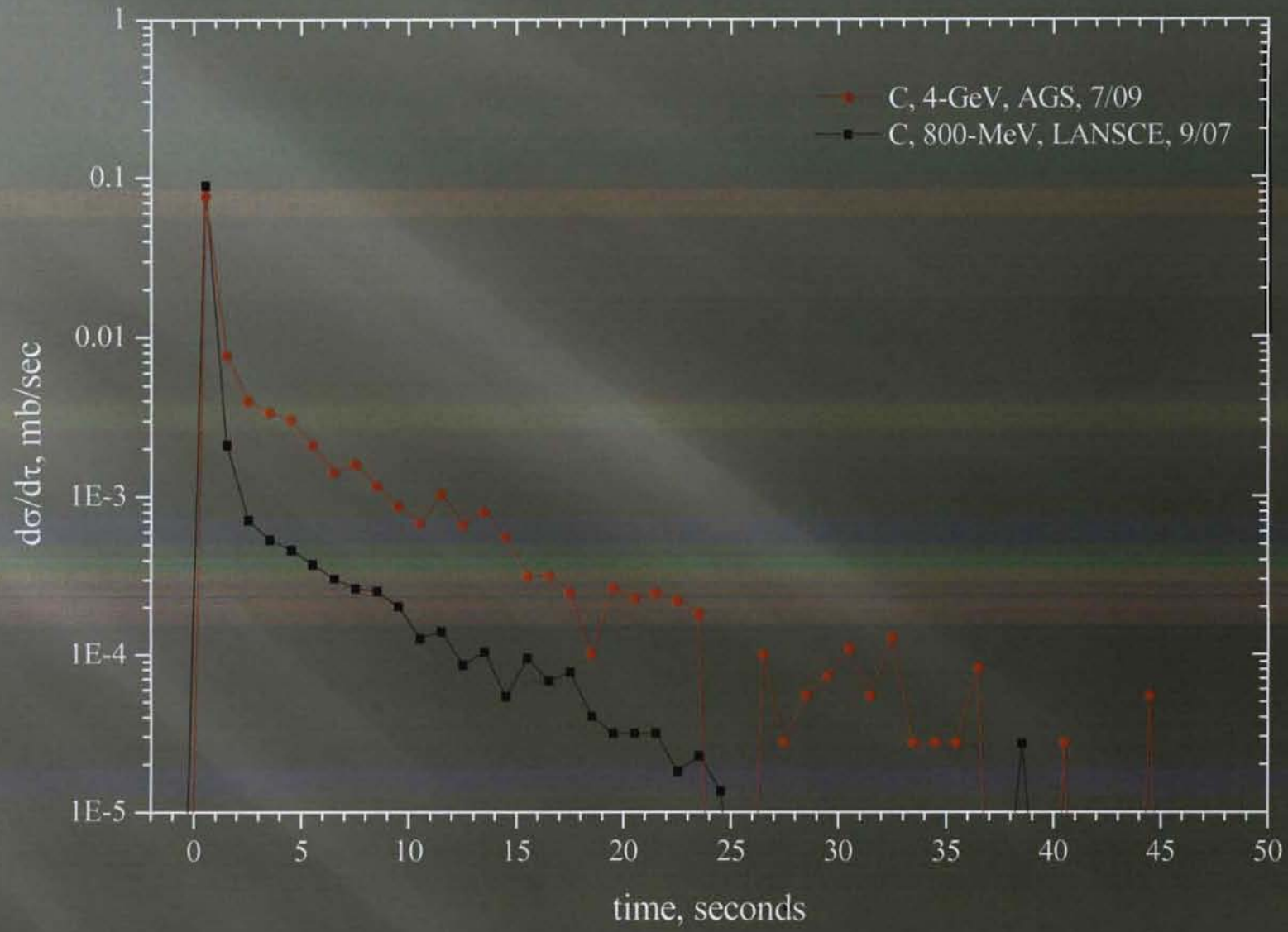
Benefits of the Surrogate Method

- Provides online measurement of scattered background component which is impossible to measure independently
- Eliminates enormous complexity involved with tracking all delayed neutron emitters in complex environment
- Flexibility: Practically any lower-group isotope can be used as a surrogate for a higher-group isotope
- η_i's fairly simple to calculate
  - Interaction probability
  - Computer code, e.g. MCNPX
- True goal is to demonstrate that backgrounds are negligible
  - Signal independent of scattered background

Delayed Neutron Cross-Section, Full-Z Comparison

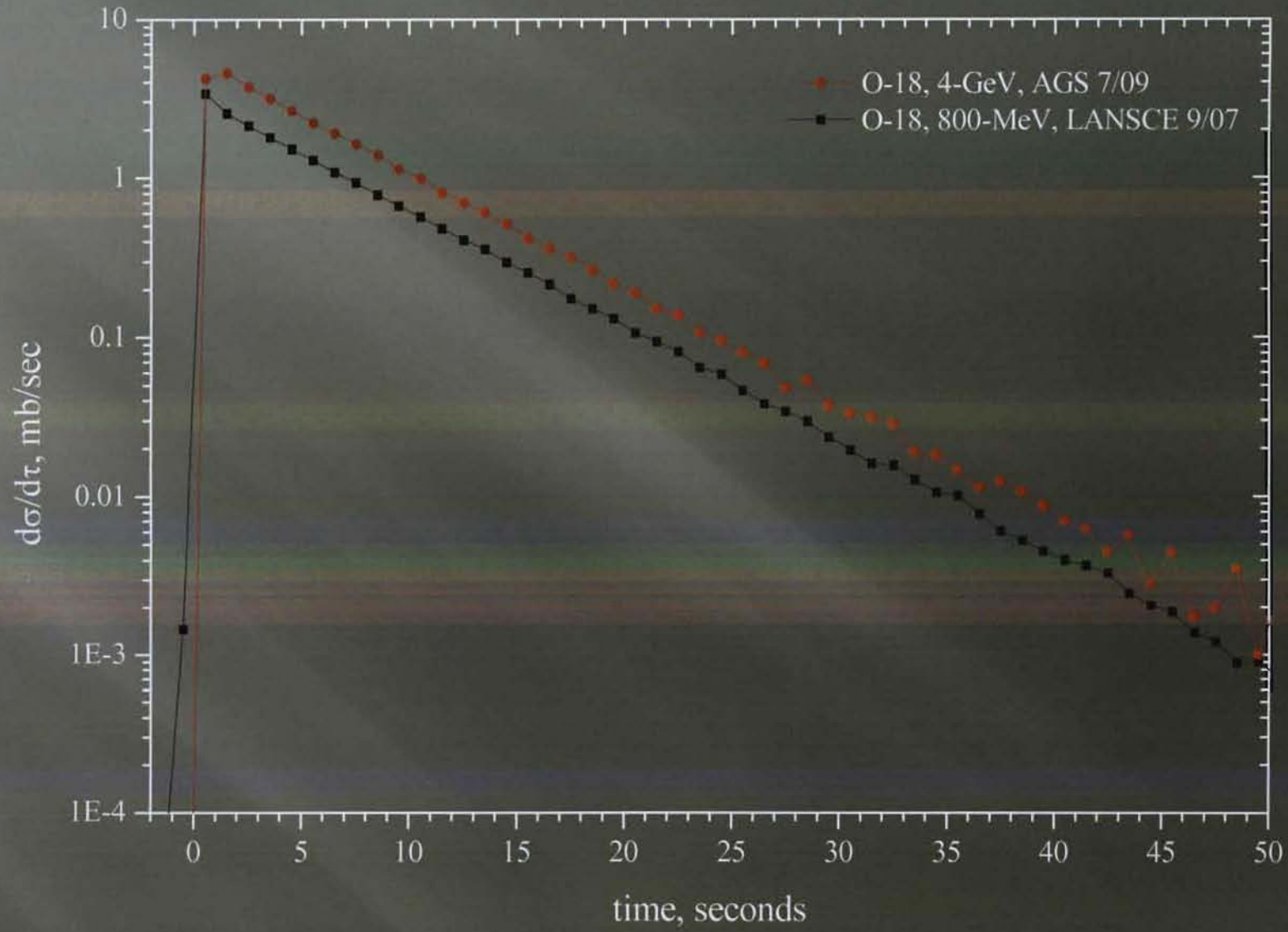


Delayed Neutron Cross-Section, Carbon Comparison



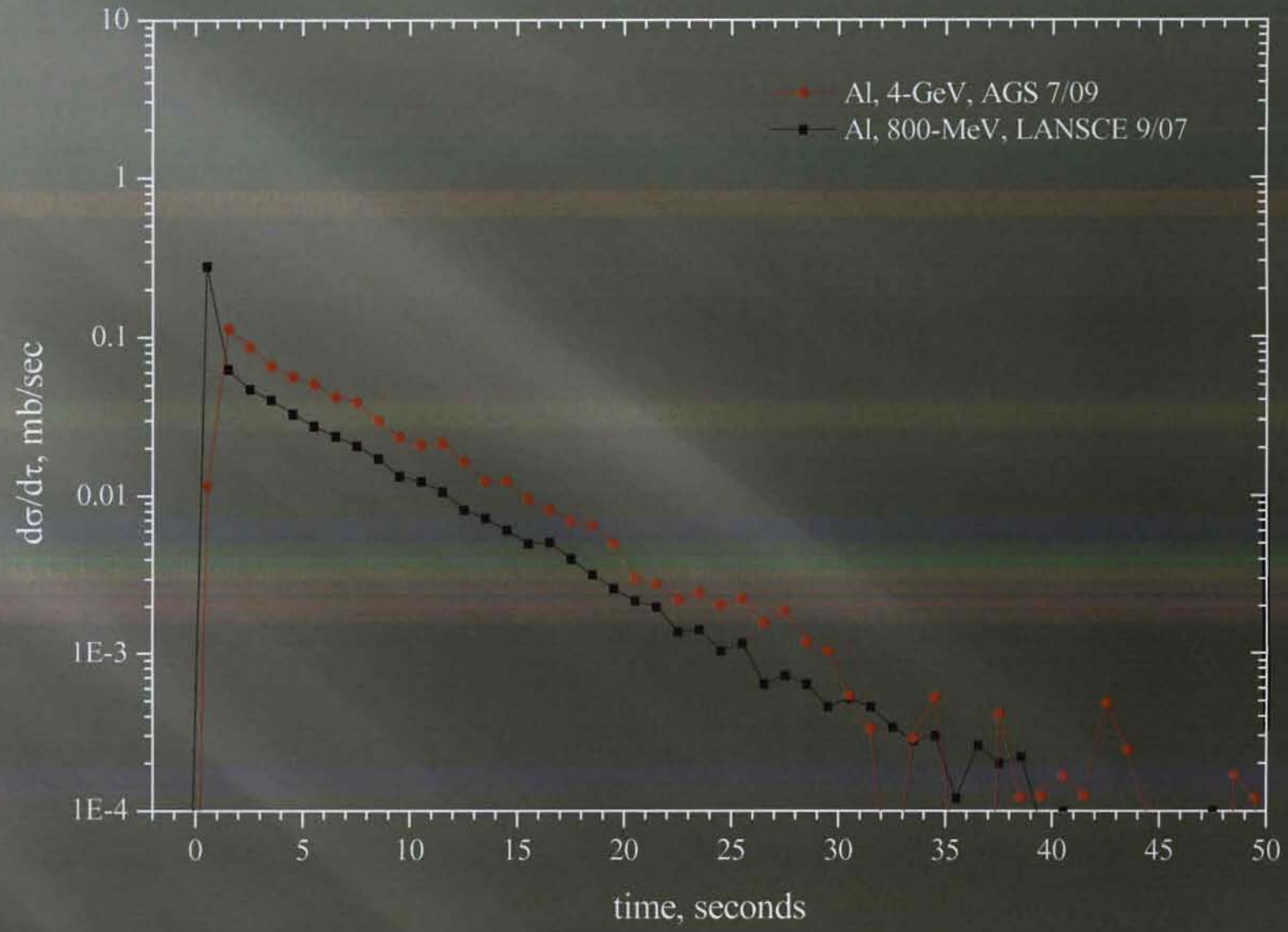
Los Alamos NATIONAL LABORATORY

Delayed Neutron Cross-Section, Oxygen-18 Comparison



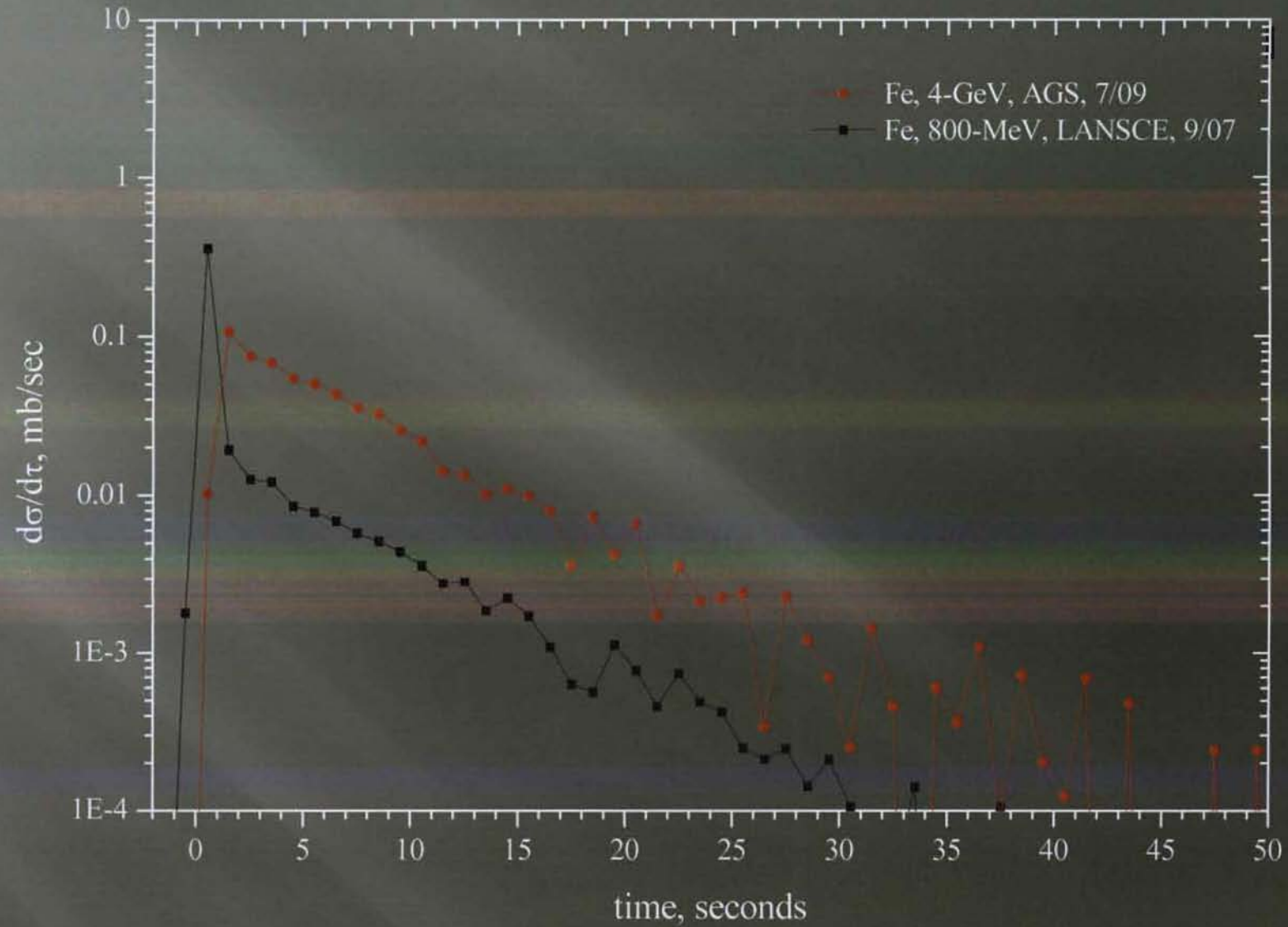
Los Alamos NATIONAL LABORATORY

Delayed Neutron Cross-Section, Aluminum Comparison



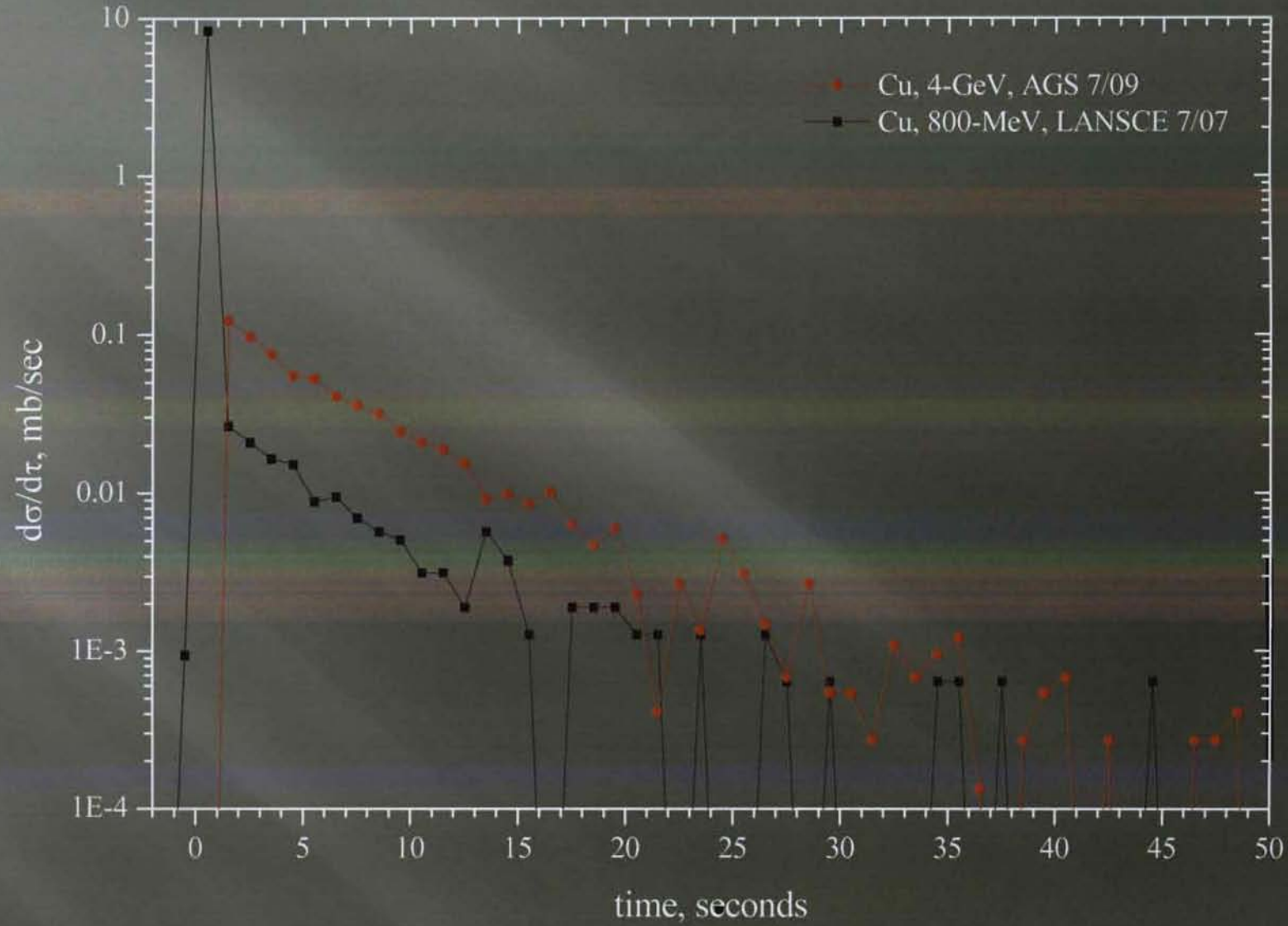
Los Alamos
NATIONAL LABORATORY

Delayed Neutron Cross-Section, Iron Comparison



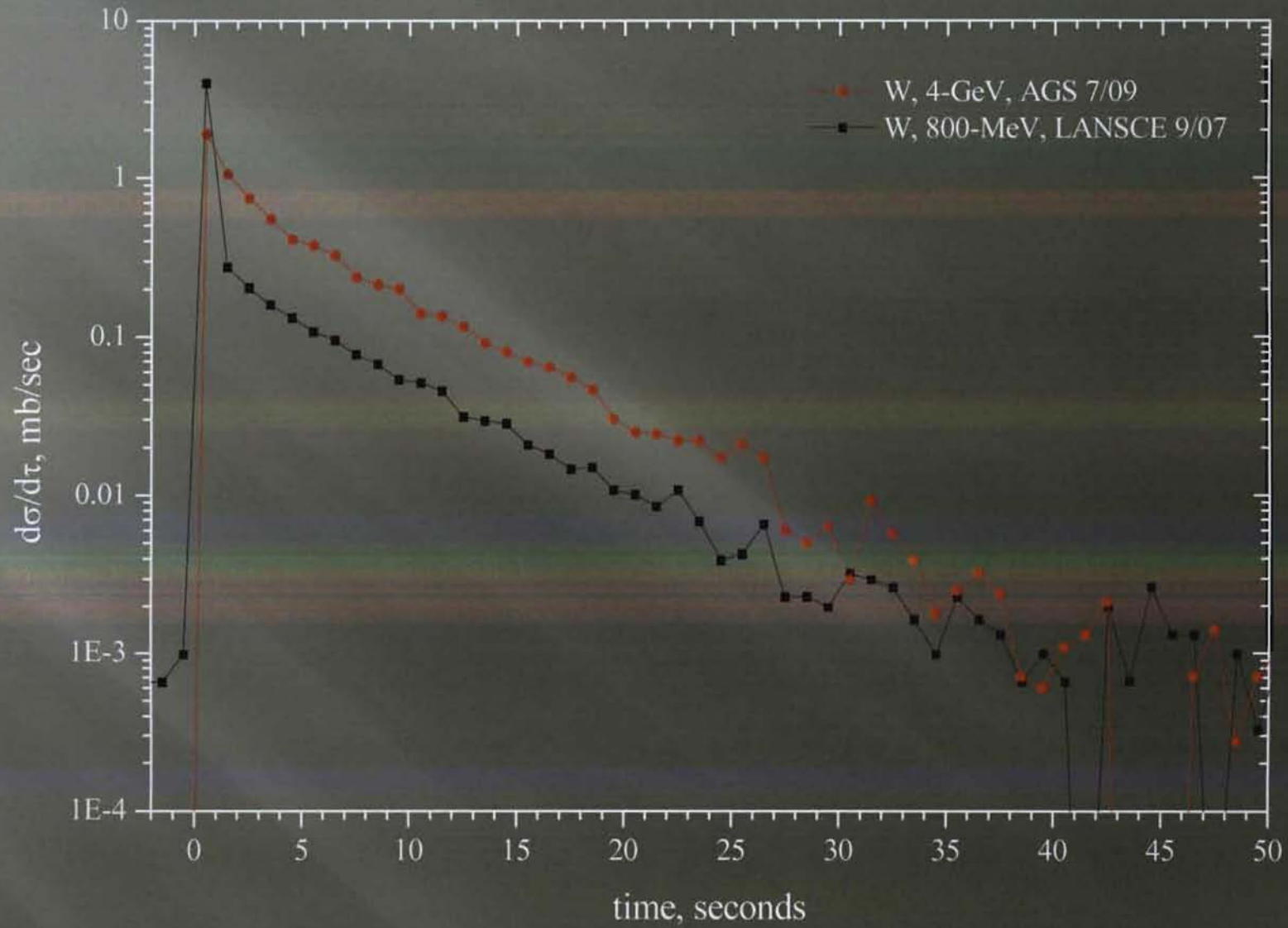
Los Alamos NATIONAL LABORATORY

Delayed Neutron Cross-Section, Copper Comparison

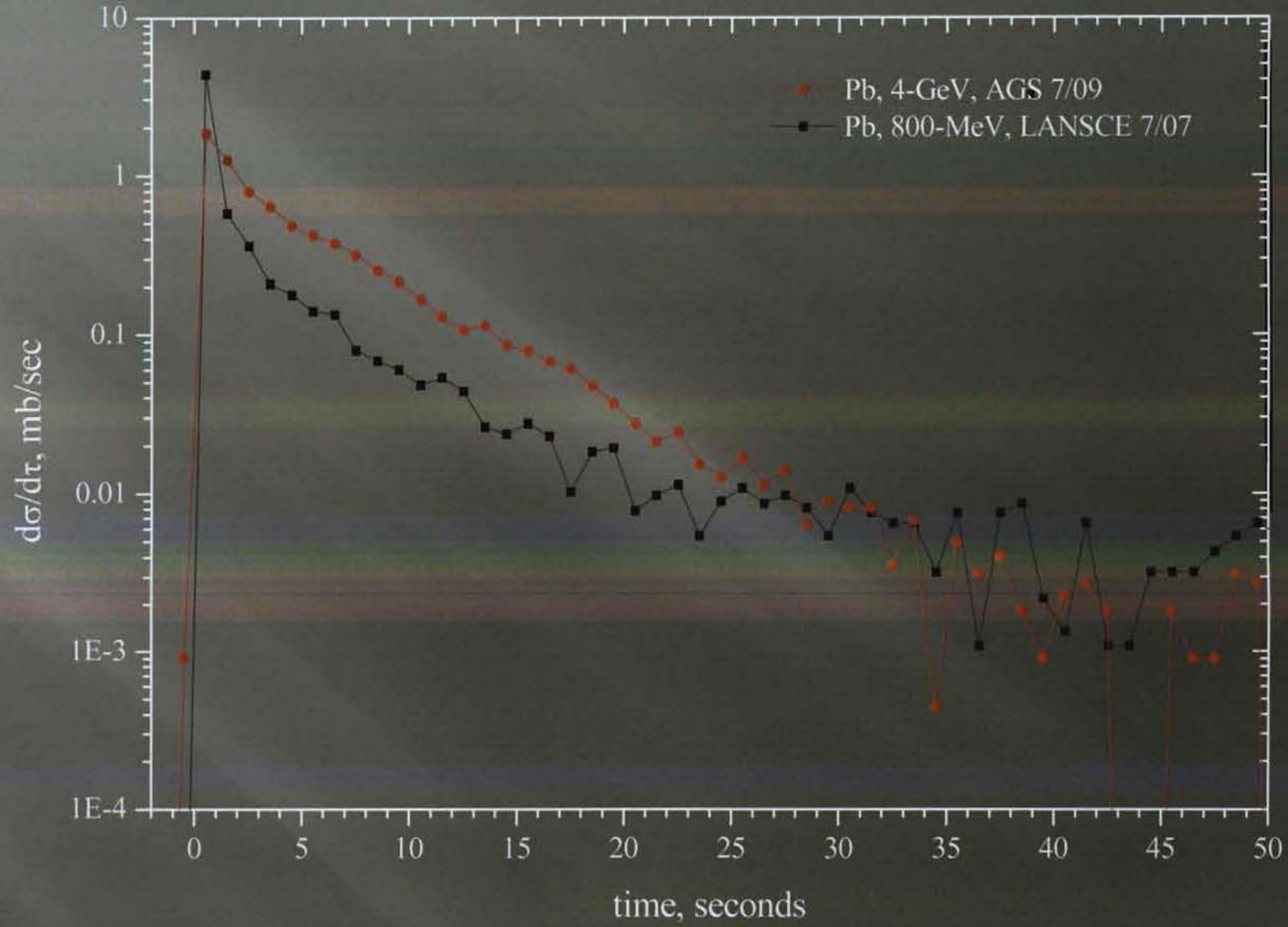


Los Alamos NATIONAL LABORATORY

Delayed Neutron Cross-Section, Tungsten Comparison

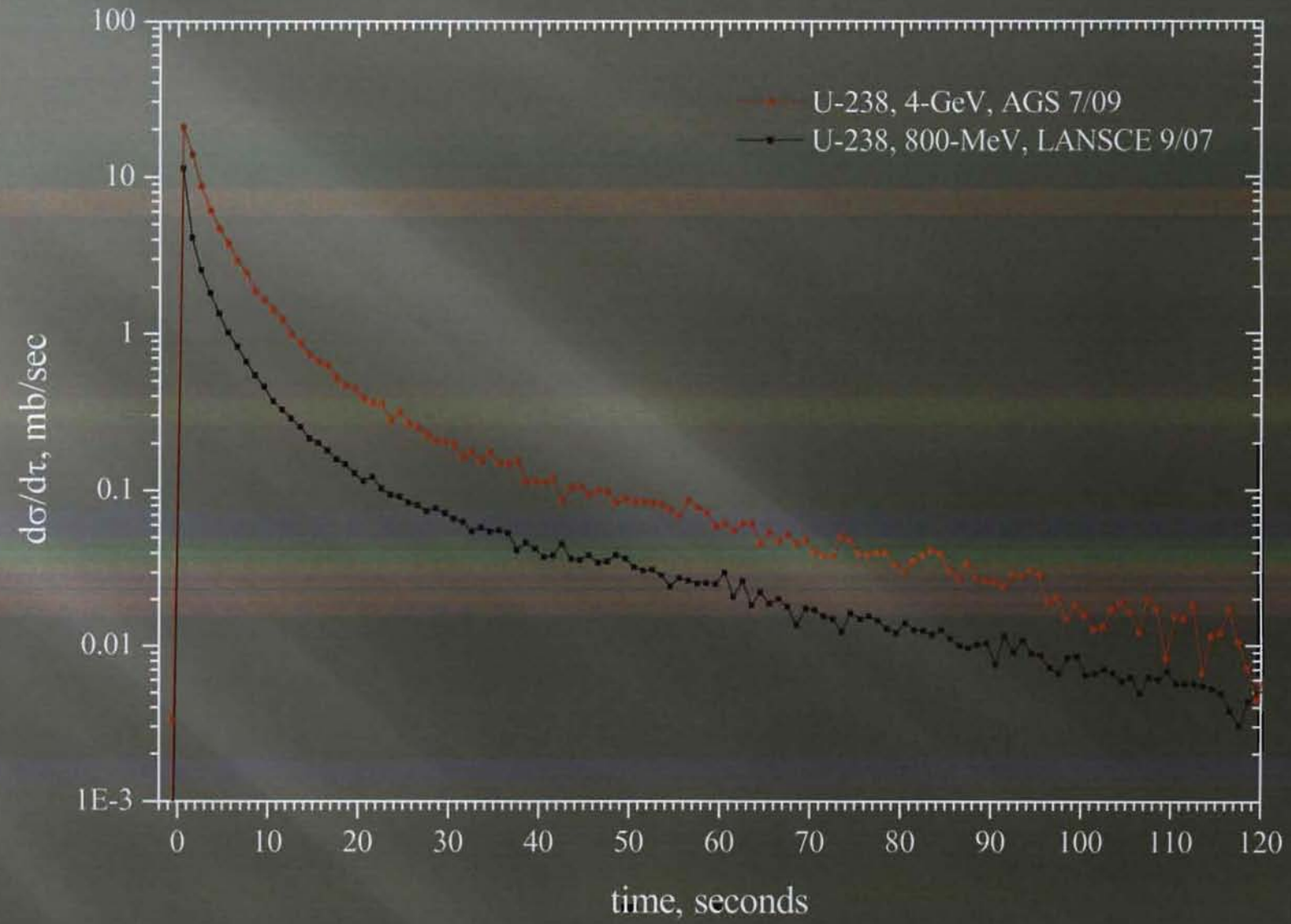


Delayed Neutron Cross-Section, Lead Comparison

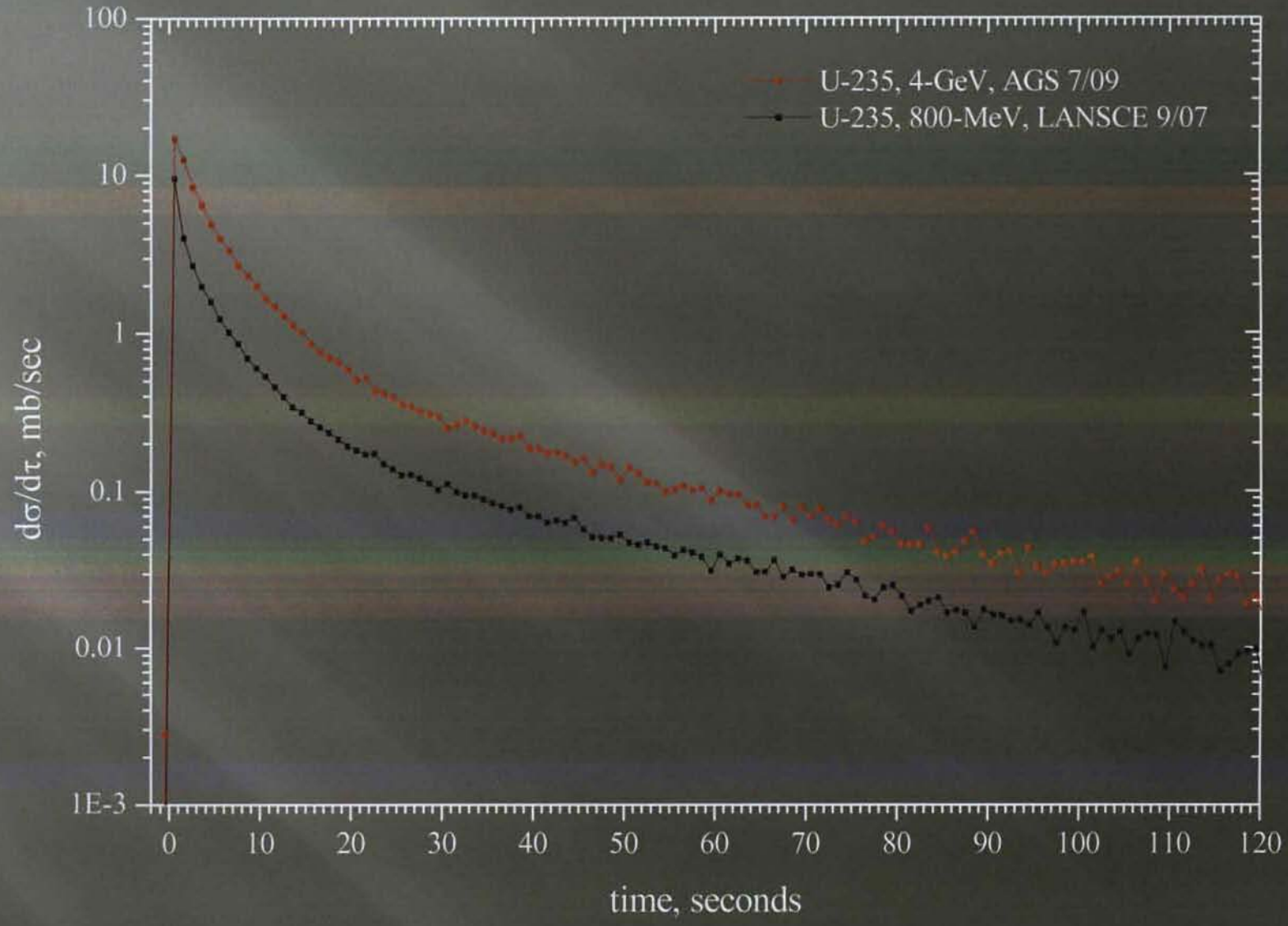


Los Alamos NATIONAL LABORATORY

Delayed Neutron Cross-Section, U-238 Comparison



Delayed Neutron Cross-Section, U-235 Comparison



Los Alamos NATIONAL LABORATORY

Conclusion and Future Work

- The proton-induced delayed neutron cross-section has been measured for several isotopes
- Signatures have been identified for fissionable materials.
  - Working on identifying fissionable vs. fissile.
- A framework is being developed to streamline benchmarking computer codes used to simulate these experiments
- Need to quantify variations in efficiency
- Need to validate surrogate method.
  - Construct DN decay curves from individual precursor yields?